

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

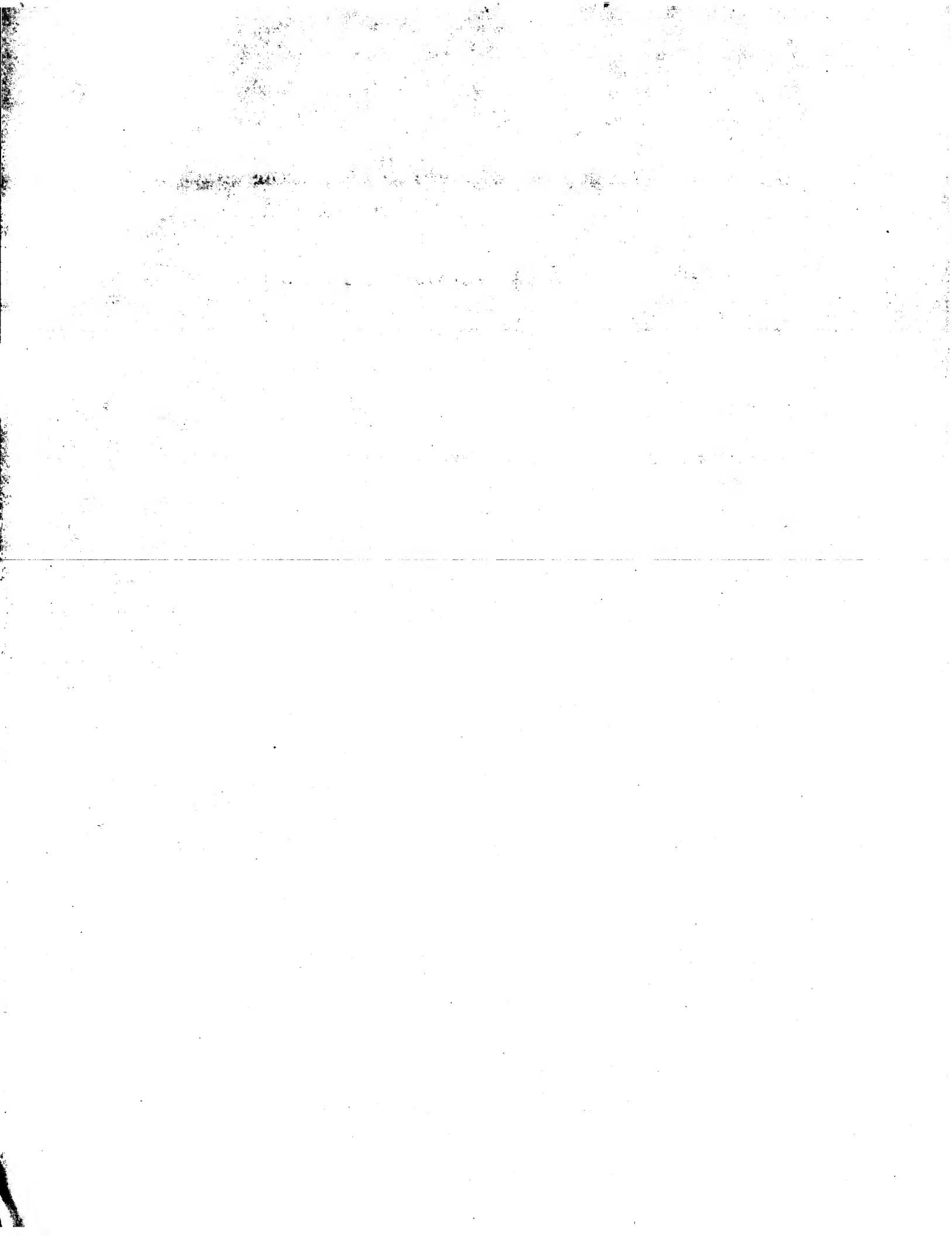
Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**





INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶ : C08J 9/04, 9/14, C09K 5/04		A1	(11) International Publication Number: WO 99/16815
			(43) International Publication Date: 8 April 1999 (08.04.99)
(21) International Application Number: PCT/US98/20507		(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).	
(22) International Filing Date: 1 October 1998 (01.10.98)		Published <i>With international search report.</i>	
(30) Priority Data: 08/941,960 1 October 1997 (01.10.97) US			
(71) Applicant: TENNECO PACKAGING INC. [US/US]; 1900 W. Field Court, Lake Forest, IL 60045 (US).			
(72) Inventor: WU, Wen, Pao; Silver Pines Drive, Pittsford, NY 14534 (US).			
(74) Agent: BLANKSTEIN, Michael, J.; P.O. Box 4433, Houston, TX 77210 (US).			

(54) Title: HIGH PRODUCTIVITY, LOW FUGITIVE VOC EMISSION FOAM PROCESS

(57) Abstract

A blowing agent for foaming polystyrene consisting essentially of at least one low permeability volatile organic compound, at least one low volatility non-volatile organic compound and, optionally, at least one high volatility non-volatile organic compound. In one embodiment, the low permeability volatile organic compound is selected from the group consisting of isopentane and isobutane, the low volatility non-volatile organic compound is acetone and the high volatility non-volatile organic compound is carbon dioxide. The blowing agent of the present invention may be used to foam a polystyrene foam structure. The blowing agent is directed to achieving low fugitive volatile organic compounds (VOC) emissions.

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece	MN	Mali	TR	Turkey
BG	Bulgaria	HU	Hungary	ML	Mongolia	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MR	Mauritania	UA	Ukraine
BR	Brazil	IL	Israel	MW	Malawi	UG	Uganda
BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
CA	Canada	IT	Italy	NE	Niger	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NL	Netherlands	VN	Viet Nam
CG	Congo	KE	Kenya	NO	Norway	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NZ	New Zealand	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	PL	Poland		
CM	Cameroon	KR	Republic of Korea	PT	Portugal		
CN	China	KZ	Kazakhstan	RO	Romania		
CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

HIGH PRODUCTIVITY, LOW FUGITIVE VOC**EMISSION FOAM PROCESS****FIELD OF INVENTION**

5 The present invention is directed to a foam process where low fugitive volatile organic compounds (VOC) emissions are achieved by employing a blowing agent. Specifically, the blowing agent comprises at least one low permeability VOC blowing agent, at least one low volatility non-VOC blowing agent and, optionally, at least one high volatility non-VOC blowing agent.

BACKGROUND OF THE INVENTION

10 Foam, such as polystyrene foam, is commonly made by combining a physical blowing agent with molten polystyrene resin under pressure and, after thorough mixing, extruding the combination through an appropriate die into a lower pressure atmosphere.

15 From about the 1950's to the present, physical blowing agents of choice have been halocarbons, hydrocarbons or combinations thereof. Examples of these include commercially available halocarbon compositions, such as dichlorodifluoromethane and trichlorofluoromethane, and the C₄ - C₆ hydrocarbons. However, these physical blowing agents have various disadvantages. For example, physical blowing agents are released into the atmosphere during and after foam production and, thus, are a source 20 of pollution. In addition, the hydrocarbon blowing agents constitute a fire hazard.

In the past, carbon dioxide has been experimented with as a blowing agent for resins, such as polystyrene resins. Carbon dioxide has been listed as a blowing agent or as a co-blowing agent in the following: United States Patent No. 3,431,164 to

Gilbert et al., United States Patent No. 4,198,363 to Noel, and United States Patent No. 4,470,938 to Johnson. Carbon dioxide has a disadvantage of a very high foaming rate caused by its high volatility. A very high foaming rate results in a high degree of corrugation in the extruded sheet. Because of the high degree of corrugation, a lower amount of blowing agent is used as compared to using a conventional hydrocarbon or halocarbon blowing agent. When a lower amount of blowing agent is used, higher viscosities and a lower output rate are obtained.

Carbon dioxide has been used as a co-blown agent or a diluent for hydrocarbon or halocarbon blowing agents. This is illustrated, for example, in United States Patent No. 4,344,710 to Johnson et al. and in an article by L.M. Zwolinski and F.J. Dwyer, Extruded Polystyrene Foam with CFC/Carbon Dioxide Blowing Agents, ANTEC'86 pages 30-33, Conference Proceedings, Society of Plastic Engineers 44th Annual Technical Conference and Exhibit. Because of the high volatility of carbon dioxide, it can only be substituted at a low level in the process. Thus, the use of carbon dioxide as a co-blown agent with hydrocarbon or halocarbons blowing agents still results in a significant amount of process emission that is a source of pollution.

Therefore, a need exists for a foam process which greatly reduces fugitive VOC emissions by reducing the concentration of the low permeability VOC blowing agent employed, while still maintaining a same extrusion output rate as a conventional process using primarily a hydrocarbon or halocarbon blowing agent.

SUMMARY OF THE INVENTION

The blowing agents of the present invention include at least one low volatility non-VOC blowing agent with at least one low permeability VOC blowing agent.

Alternatively, at least one high volatility non-VOC blowing agent may be added to the low volatility non-VOC blowing agent and the low permeability VOC blowing agent. The term VOC, as used herein, is defined by the Environmental Protection Agency (EPA) as any compound of carbon, excluding carbon monoxide, carbon dioxide, 5 carbonic acid, metallic carbides or carbonates, and ammonium carbonate, which participates in atmospheric photochemical reactions. This definition of VOC, including a list of excluded VOCs, is illustrated, for example, in "Proposed Rules of the Environmental Protection Agency" by Carol M. Browner in the Federal Register at Vol. 62, No. 164 on August 25, 1997 which is incorporated herein by reference in its 10 entirety.

In one embodiment, the low volatility non-VOC blowing agent is acetone, the low permeability VOC blowing agent is a hydrocarbon (such as isopentane) or a halocarbon, and the high volatility non-VOC blowing agent is carbon dioxide.

The blowing agent of the present invention may be used with polystyrene to 15 form a polystyrene foam structure.

BRIEF DESCRIPTION OF THE DRAWINGS

Other objects and advantages of the invention will become apparent upon reading the following detailed description and upon reference to the drawings in which

20 FIGURE is a schematic flow diagram of an overall sequence of operations according to one embodiment involved in the manufacture of a foamed polystyrene sheet with the blowing agents of the present invention.

While the invention is susceptible to various modifications and alternative forms, a specific embodiment thereof has been shown by way of example in the

drawing and will herein be described in detail. It should be understood, however, that it is not intended to limit the invention to the particular forms disclosed, but on the contrary, the intention is to cover all modifications, equivalents, and alternatives falling within the spirit and scope of the invention as defined by the appended claims.

5

DESCRIPTION OF ILLUSTRATIVE EMBODIMENTS

The foam process of the present invention employs a blowing agent to achieve low fugitive VOC emissions. The blowing agent comprises at least one low permeability VOC blowing agent, at least one low volatility non-VOC blowing agent and, optionally, at least one high volatility non-VOC blowing agent.

10

Of the thermoplastic resins which can be foamed in accordance with the present invention, the styrene homopolymers and copolymers containing a predominant portion of styrene, *i.e.*, greater than about 50 weight percent styrene are preferred with those containing greater than about 75 weight percent styrene being especially preferred. With respect to a styrene copolymer, the comonomer can be any other ethylenically unsaturated material such as the conjugated 1,3-dienes, *e.g.*, butadiene, isoprene, etc., alpha-beta-unsaturated monocarboxylic acids and derivatives thereof, *e.g.*, acrylic acid, methyl acrylate, ethyl acrylate, butyl acrylate, 2-ethylhexyl acrylate, and the corresponding esters of methacrylic acid, acrylamide, methacrylamide, acrylonitrile, methacrylonitrile, etc. If desired, blends of a styrene polymer with other polymers can be employed, *e.g.*, blends of a styrene polymer with polyphenylene oxide. In any of the foregoing polymers, all or a portion of the styrene can be replaced with one or more closely related homologues of styrene such as alpha-methylstyrene, o-, m-, and p-methylstyrene, o-, m-, p-ethylstyrene, 2, 4-dimethylstyrene, etc.

In the present invention, at least one of the VOC blowing agents exhibits low permeability in the polymer being foamed. A low permeability blowing agent is defined as having a permeability lower than about 1×10^{-12} cc(STP)(cm)/[(sec)(cm²)(cmHg)] and preferably lower than about 5×10^{-13} cc(STP)(cm)/[(sec)(cm²)(cmHg)]. In the case of polystyrene, the preferred low permeability VOC blowing agents are cyclopentane, isopentane, isobutane, 2-2 dimethyl butane, cyclohexane and mixtures thereof.

The addition of at least one low volatility non-VOC blowing agent further reduces the viscosity of the melt to the same degree as the hydrocarbon VOC blowing agent originally employed. A low volatility blowing agent is defined as having a boiling point higher than ambient temperature, e.g. about 23°C, and preferably having a boiling point greater than about 40°C. One example of a low volatility non-VOC blowing agent that may be used in the present invention is acetone. In addition, acetone may be combined with water to comprise a low volatility non-VOC blowing agent. Another example of a potential low volatility non-VOC is methyl acetate. Currently, methyl acetate is classified as a VOC blowing agent, however, methyl acetate may be reclassified as a non-VOC blowing agent under a proposal to the Environmental Protection Agency (EPA). If methyl acetate is classified as a non-VOC blowing agent, it is contemplated that it could be mixed or co-injected with acetone or an acetone/water mixture.

One example of a high volatility non-VOC blowing agent that in the present invention is carbon dioxide (CO₂). In one embodiment of the present invention, carbon dioxide is in the liquid state before injection into the extruder. However, carbon dioxide in gaseous state or supercritical fluid state may also be used in the

present invention. It is contemplated that other high volatility non-VOC blowing agents like nitrogen, ethane and hydrofluorocarbons, such as HFC-152a, may be employed. It is additionally contemplated that the above high volatility non-VOC blowing agents may be mixed. A high volatility blowing agent is defined as having a
5 boiling point below about 0°C, and preferably below about -20°C.

The total amount of blowing agent used will depend on conditions such as extrusion process conditions at mixing, the composition of the extrudate, and the desired density of the foamed sheet. The remainder of the foamed sheet density is comprised of a thermoplastic resin and any additives that may be used including, but
10 not limited to, foaming additives and colorants. For a foamed sheet density of from about 1 to about 15 lb./ft³, typically the total weight percentage of the blowing agent will be from about 2 to about 10 weight percent of the extrudate. The extrudate is defined as including a blowing agent, a polystyrene or the like, and any additives,
15 colorants, etc. The preferred range is from about 2 to about 8 wt.% blowing agent, and more preferably from about 2 to about 6 wt.% blowing agent of the extrudate.

Specifically, the low permeability VOC comprises from about 0.5 to about 5 wt.%, and preferably from about 1 to about 3 wt.% of the extrudate. The low volatility non-VOC comprises from about 0.25 to about 5 wt.%, and preferably from about 0.5 to about 3 wt.% of the extrudate. The high volatility non-VOC comprises
20 from about 0.25 to about 5 wt.%, and preferably from about 0.5 to about 3 wt.% of the extrudate. It is preferred that the amount of the high volatility non-VOC blowing agent be maintained at a certain amount in the system to achieve better final sheet density control.

A nucleating agent or combination of such agents may be employed in the present invention for advantages such as their capability for regulating cell formation and morphology. Known nucleating agents such as talc, mixtures of sodium bicarbonate and citric acid, and the like may be employed in the present invention.

5 If desired, fillers, colorants, light and heat stabilizers, plasticizers, chemical blowing agents, foaming additives and plastic compounding agents can be included in the foamable resin compositions.

A conventional two-extruder tandem system may be used for extruding the foam sheet of the present invention. A single extruder with proper cooling may also 10 be employed.

For example, polystyrene resin pellets are admixed with a nucleating agent such as talc. These materials are continuously fed into a hopper of a 4 ½ inch diameter screw primary extruder. The feed mixture is conveyed forward by the helical screw within a barrel of the extruder as the mixture is mixed, compressed, heated, and 15 converted to molten form. The conversion to molten form occurs prior to reaching the injection zone where the blowing agent is added. The blowing agent of the present invention is injected into the polystyrene composition at a point where the polystyrene is in a molten condition (i.e. beyond the feed zone).

After injecting the blowing agent, the mixture is continuously mixed in the 20 primary extruder at pressure high enough to ensure a homogeneous solution of the resin and the blowing agent mixture. The molten mixture is then conveyed into a cooled secondary extruder where the mixture is passed along the length of the secondary extruder at low shear. During this time, the molten mixture is cooled and

additional mixing take place. The mixture is then extruded through a die into a lower pressure zone such as ambient pressure.

It is desirable that the melt temperature of the polymer entering the die be reduced to a relatively low value. This lower melt temperature of the polymer 5 entering the die produces a foam sheet with an improved property balance between the machine direction (MD) and the transverse direction (TD).

With reference to the FIGURE, a two-extruder tandem system 10 may be used for extruding the foam sheet of the present invention. Polystyrene resin pellets are mixed with additive(s), including a nucleating agent, to form a feed mixture which is 10 fed continuously into a hopper 11 of a 4 ½ inch diameter screw primary extruder 13. The feed mixture is conveyed forward by a helical screw within a barrel of the extruder as the feed mixture is mixed, compressed, heated and melted prior to reaching the injection zone for adding a blowing agent. The blowing agent is added at point 15. Thus, the blowing agent of the present invention is injected into the 15 polystyrene additives mixture (feed mixture) at a point beyond the feed zone where the polystyrene is melted.

Following injection of the blowing agent, the mixture is continuously mixed in the primary extruder 13 and subsequently passed, at a high enough pressure that the blowing agents remain in solution, through a hollow adapter section 17 into a cooled 6 inch diameter secondary tandem extruder 19. The molten mixture is passed along the 20 length of the cooled secondary extruder at low shear where cooling and additional mixing occur. The mixture is then expressed through an annular die 21. The foamable polystyrene polymer is extruded through the annular die 21 in the form of an elongated bubble or tube 23 which is expanded and drawn over a cylindrical surface

of a cooling and sizing drum 25 and slit to form sheet stock 27. The sheet stock 27 is taken up on one or more winding reels 29.

In the FIGURE, a thermoplastic resin, *e.g.*, a polystyrene homopolymer, containing a nucleating agent such as talc is introduced into the primary extruder 13. 5 The primary extruder 13 melts the mixture and then combines the mixture with the blowing agent of the present invention under pressure. The operating conditions of the primary extruder 13, for example, include an extruder exit pressure of from about 1000 to about 10,000 psi, and preferably from about 2,000 to about 5,000 psi. The temperature of the primary extruder 13 ranges from about 380 to about 550 °F, and 10 preferably from about 420 to about 500 °F.

The extrudate from the primary extruder 13 is thereafter introduced to the secondary extruder 19. The operating conditions of the secondary extruder 19, for example, include an extruder exit pressure of from about 1,000 to about 6,000 psi, and preferably from about 1,500 to about 3,500 psi. The temperature of the extrudate 15 from the secondary extruder 19 can range from about 200 to about 360°F, and preferably from about 250 to about 340°F.

The foam sheet resulting from the foam tube, extruded in accordance with the invention, can vary from about 0.03 to about 1.0 inch or more in thickness, *e.g.*, from about 0.03 to about 0.5 inch in thickness. The resulting foam sheet has a density of 20 from about 1 to about 15 lb/ft³, *e.g.*, from about 2.0 to about 9.0 lb/ft³. When the foam sheet is extruded to a thickness of from about 0.25 inch to 0.50 inch or more, it is suitable for use as an insulation board.

Example

The blowing agents were tested with the results shown in the Table. All of the Inventive Foams 2-5 were comprised of various amounts of a low permeability VOC blowing agent (isopentane), a high volatility non-VOC blowing agent (CO_2), and a low volatility non-VOC blowing agent (acetone). Comparative Foam 1 comprised a 5 low permeability VOC blowing agent (isopentane) and a high volatility non-VOC blowing agent (CO_2) without a low volatility non-VOC blowing agent.

In producing Comparative Foam 1, high heat general purpose polystyrene (GPPS) was melted in a 2-1/2" extruder. The blowing agents (isopentane and carbon dioxide) were injected into a first extruder at a point after a first metering zone of the 10 first extruder. The polystyrene/blowing agent mixture was then homogenized in the remainder of the first extruder. This mixture was then conveyed through a second extruder and cooled to about 300°F after which it was extruded through an annular die to make a closed cell polystyrene foam at a rate of 139 lb/hr. The input amount of 15 isopentane was controlled to 4.2 wt.% of the extrudate. Similarly, the input amount of carbon dioxide was controlled to 0.5 wt.% of the extrudate. The sheet of Comparative 15 Foam 1 had an extruded density of 3.8 lb/ft³, which is considered typical for polystyrene food packaging foam.

The sheet of Comparative Foam 1 aged for 7 days after which parts were formed having a final density of 2.2 lb/ft³. The amount of isopentane remaining in the 20 foam parts was measured at regular intervals using gas chromatography. The measurements were made in accordance with the procedure set forth in the article entitled "Gas Chromatographic Determination of Residual Blowing Agents in Polystyrene Foams" to Krutchen, C.M. and W.P. Wu, Proceeding SPE ANTEC '88, pp. 704-706 (1988). The amount of isopentane remaining in the foam parts 28 days

after extrusion was 2.98%. The difference between the 4.2% input and the 2.98% retained isopentane (1.22%) was the blowing agent loss to the atmosphere.

All of the Inventive Foams 2-5 were made under similar conditions except for the specific amounts of isopentane and carbon dioxide. In addition, Inventive Foams 5 2-5 included acetone in its blowing agent mixture.

Referring to the Inventive Foams 2-5, the weight percent of the isopentane was in the range of from 0.8 wt.% to 1.88 wt.%, the weight percent of the acetone was in the range of from 1.88 wt.% to 3.1 wt.%, and the weight percent of the carbon dioxide was in the range from 0.60 wt.% to 0.73 wt.%. All these above weight percentages 10 were based on the extrudate. The remainder of the feed mixture comprised a high heat general purpose polystyrene.

Referring to the Table, the Inventive Foams 2-5 showed improvement in the blowing agent loss in 30 days over the Comparative Foam 1. Specifically, when the isopentane level was reduced to about 1 to about 2 wt.%, very little permeated out of 15 the foam sheet after an extended period of time (see blowing agent loss in 30 days in Inventive Foams 2-5). Thus, the Inventive Foams 2-5 exhibited low VOC emissions. Additionally, the blowing agents of Inventive Foams 2-5 had an equal or better extrusion rate than Comparative Foam 1 (see rate in lb/hr in Table).

The foamed sheet produced in accordance with this invention is entirely 20 suitable for thermoforming. One such example of a thermoforming process and apparatus is described in United States Patent No. 4,485,059, which is incorporated herein by reference in its entirety. Employing this or other thermoforming techniques, a variety of useful articles such as cartons, containers, cups, lids, bowls, plates, trays, etc., can be manufactured with great production efficiency and low cost. The foamed

sheet manufactured with the blowing agent of the present invention is also suitable for the preparation of insulation board, wherein the insulating boards are extruded and foamed to a predetermined size.

While the present invention has been described with reference to one or more particular embodiments, those skilled in the art will recognize that many changes may be made thereto without departing from the spirit and scope of the present invention. Each of these embodiments and obvious variations thereof is contemplated as falling within the spirit and scope of the claimed invention, which is set forth in the following claims.

TABLE

	Comparative Foam #1	Inventive Foam #2	Inventive Foam #3	Inventive Foam #4	Inventive Foam #5
isopentane (wt%)	4.2%	1.88%	1.47%	1.07%	0.80%
acetone (wt%)	0%	1.88%	2.21%	2.76%	3.1%
CO ₂ (wt%)	0.50%	0.73%	0.69%	0.72%	0.60%
rate (lb/hr)	139	146	151	147	142
part density (lb/ft ³)	2.22	3.13	2.50	2.72	2.83
VOC Blowing Agent loss (wt% in 28-30 days)	1.22%	0.18%	0.02%	0.14%	0.03%

WHAT IS CLAIMED IS:

1. A blowing agent for foaming polystyrene consisting essentially of: at least one low permeability compound, said low permeability compound having a permeability in said polystyrene lower than about 1×10^{-12} cc(STP)(cm)/ and being 5 classified as a volatile organic compound (VOC) by the Environmental Protection Agency(EPA);
at least one low volatility compound, said low volatility compound having a boiling point higher than about 23°C and not being classified as a VOC by the EPA;
and, optionally, at least one high volatility compound, said high volatility 10 compound having a boiling point lower than about 0°C and not being classified as a VOC by the EPA;
wherein said blowing agent comprises less than about 42 wt.% of said low permeability compound.
2. The blowing agent of claim 1 wherein at least one of said low permeability 15 compound is selected from the group consisting of isopentane, isobutane, and mixtures thereof, at least one of said low volatility compound is acetone and at least one of said high volatility compound is carbon dioxide.
3. The blowing agent of claim 1 wherein said blowing agent comprises less than 34 wt.% of said low permeability compound.
- 20 4. The blowing agent of claim 1 wherein at least one low permeability compound is selected from the group consisting of cyclopentane, isopentane, isobutane, 2-2 dimethyl butane, cyclohexane and mixtures thereof.
5. The blowing agent of claim 1 wherein at least one low volatility compound is selected from the group consisting of acetone, an acetone/water mixture, methyl 25 acetate, and mixtures thereof.
6. The blowing agent of claim 1 wherein said at least one high volatility compound is selected from the group consisting of carb dioxide, nitrogen, hydrofluorocarbons, ethane and mixtures thereof.

7. A polystyrene foam structure prepared by the process comprising the steps of:
- a) melting a mixture comprising a thermoplastic polymer;
 - b) dissolving an effective amount of blowing agent in the mixture of step 5 (a), said blowing agent consisting essentially of at least one low permeability compound, at least one low volatility compound and, optionally, at least one high volatility compound, said low permeability compound having a permeability in said polystyrene foam structure lower than about 1×10^{12} cc(STP)(cm)/ and being classified as a volatile organic compound (VOC) by the Environmental Protection Agency (EPA), said low volatility compound having a boiling point higher than about 10 23°C and not being classified as a VOC by the EPA, said high volatility compound having a boiling point lower than about 0°C and not being classified as a VOC by the EPA;
 - (c) forming an extrudate, said extrudate comprising from about 0.5 to 15 about 3 weight percent low permeability compound;
 - (d) transferring the extrudate to an expansion zone; and
 - (e) permitting the extrudate to expand in said expansion zone to produce said polystyrene foam structure;
wherein said polystyrene foam structure has a density from about 1 to about 15 20 lb/ft³.
8. The polystyrene foam structure of claim 7 wherein at least one of said low permeability compound is selected from the group consisting of isopentane, isobutane and mixtures thereof, at least one of said low volatility compound is acetone and at least one of said high volatility compound is carbon dioxide.
- 25 9. The polystyrene foam structure of claim 7 wherein said extrudate comprises from about 2 to about 10 weight percent blowing agent.
10. The polystyrene foam structure of claim 9 wherein said extrudate comprises from about 3 to about 8 weight percent blowing agent.
- 30 11. The polystyrene foam structure of claim 10 wherein said extrudate comprises from about 4 to about 6 weight percent blowing agent.

12. The polystyrene foam structure of claim 7 wherein said extrudate comprises from about 0.5 to about 1.88 wt.% of said low permeability compound.

13. The polystyrene foam structure of claim 7 wherein said extrudate comprises from about 1 to about 3 wt.% of said low permeability compound.

5 14. The polystyrene foam structure of claim 7 wherein said low volatility non-VOC comprises from about 0.25 to about 5 wt.% of the extrudate.

15. The polystyrene foam structure of claim 14 wherein said extrudate comprises from about 0.5 to about 3 wt.% said low volatility compound.

10 16. The polystyrene foam structure of claim 7 wherein said extrudate comprises from about 0.25 to about 5 wt.% said high volatility compound.

17. The polystyrene foam structure of claim 16 wherein said extrudate comprises from about 0.5 to about 3 wt.% said high volatility compound.

15 18. The polystyrene foam structure of claim 7 wherein at least one low permeability compound is selected from the group consisting of cyclopentane, isopentane, isobutane, 2-2 dimethyl butane, cyclohexane and mixtures thereof.

19. The polystyrene foam structure of claim 7 wherein at least one low volatility compound is selected from the group consisting of acetone, an acetone/water mixture, methyl acetate, and mixtures thereof.

20 20. The polystyrene foam structure of claim 7 wherein at least one high volatility compound is selected from the group consisting of carbon dioxide, nitrogen, hydrofluorocarbons, ethane and mixtures thereof.

21. A process for producing a polystyrene foam structure comprising the steps of:

- 25 (a) melting mixture comprising a thermoplastic polymer;
(b) dissolving an effective amount of blowing agent in the mixture of step (a), said blowing agent consisting essentially of at least one low permeability compound, at least one low volatility compound and, optionally, at least one high volatility

compound said low permeability compound having a permeability in said polystyrene foam structure lower than about 1×10^{12} cc(STP)(cm)/ and being classified as a volatile organic compound (VOC) by the Environmental Protection Agency(EPA), said low volatility compound having a boiling point higher than about 23°C and not being 5 classified as a VOC by the EPA, said high volatility compound having a boiling point lower than about 0°C and not being classified as a VOC by the EPA;

- (c) forming an extrudate, said extrudate comprising from about 0.5 to about 3 wt.% low permeability compound;
- (d) transferring the extrudate to an expansion zone; and
- 10 (e) permitting the extrudate to expand in said expansion zone to produce said polystyrene foam structure, said polystyrene foam structure having a density from about 1 to about 15 lb/ft³.

22. The process for producing a polystyrene foam structure of claim 21 wherein at least one of said low permeability compound is selected from the group 15 consisting of isopentane, isobutane and mixtures thereof, at least one of said low volatility compound is acetone and at least one of said high volatility compound is carbon dioxide.

23. The process for producing a polystyrene foam structure of claim 21 wherein at least one of said low volatility compound is selected from the group 20 consisting of acetone, an acetone/water mixture, methyl acetate and mixtures thereof.

24. The process for producing a polystyrene structure of claim 21 wherein at least one of said low permeability compound is selected from the group consisting of cyclopentane, isopentane, isobutane, 2-2 dimethyl butane, cyclohexane and mixtures thereof.

25. The process for producing a polystyrene structure of claim 21 wherein at least one of said high volatility compound is selected from the group consisting of carbon dioxide, nitrogen, hydrofluorocarbons, ethane and mixtures thereof.

26. The process for producing a polystyrene structure of claim 21 wherein said extrudate comprises from about 0.5 to about 1.88 wt.% low permeability compound.

27. The process for producing a polystyrene structure of claim 21 wherein
5 said extrudate comprises from about 0.25 to about 5 wt.% low volatility compound.

28. The process for producing a polystyrene structure of claim 27 wherein said extrudate comprises from about 0.5 to about 3 wt.% low volatility compound.

29. The process for producing a polystyrene structure of claim 21 wherein said extrudate comprises from about 0.25 to about 5 wt.% high volatility compound.

10 30. The process for producing a polystyrene structure of claim 29 wherein said extrudate comprises from about 0.5 to about 3 wt.% high volatility compound.

31. The process for producing a polystyrene structure of claim 21 wherein said extrudate comprises from about 2 to about 10 wt.% blowing agent.

32. The process for producing a polystyrene structure of claim 31 wherein
15 said extrudate comprises from about 3 to about 8 wt.% blowing agent.

33. The process for producing a polystyrene structure of claim 32 wherein said extrudate comprises from about 4 to about 6 wt.% blowing ag~~e~~.

34. A process for producing a polystyrene foam structure comprising the steps of:

- 20 (a) melting a mixture comprising a thermoplastic polymer;
- (b) dissolving an effective amount of blowing agent in the mixture of step (a), said blowing agent consisting essentially of at least one low permeability compound, at least one low volatility compou~~nd~~ and, optionally, at least one high volatility compound, said low permeability compound being selected from the group

consisting cyclopentane, isopentane, isobutane, 2-2 dimethyl butane, cyclohexane and mixtures thereof, said low volatility compound being selected from the group consisting of acetone, an acetone/water mixture, methyl acetate and mixtures thereof, said high volatility compound being selected from the group consisting of carbon dioxide, nitrogen, hydrofluorocarbons, ethane and mixtures thereof;

- (c) forming an extrudate, said extrudate comprising from about 0.5 to about 3 wt.% low permeability compound;
- (d) transferring the extrudate to an expansion zone; and
- (e) permitting the extrudate to expand in said expansion zone to produce 10 said polystyrene foam structure, said polystyrene foam structure having a density from about 1 to about 15 lb./ft³.

35. The process for producing a polystyrene foam structure of claim 34 wherein at least one of said low permeability compound is selected from the group consisting of isopentane, isobutane and mixtures thereof, at least one of said low 15 volatility compound is acetone and at least one of said high volatility compound is carbon dioxide.

36. The process for producing a polystyrene structure of claim 34 wherein said extrudate comprises from about 0.5 to about 1.88 weight percent low permeability compound.

37. The process for producing a polystyrene structure of claim 34 wherein 20 said extrudate comprises from about 0.25 to about 5 wt.% low volatility compound.

38. The process for producing a polystyrene structure of claim 37 wherein said extrudate comprises from about 0.5 to about 3 wt.% low volatility compound.

39. The process for producing a polystyrene structure of claim 34 wherein said extrudate comprises from about 0.25 to about 5 wt.% high volatility compound.

40. The process for producing a polystyrene structure of claim 39 wherein
5 said extrudate comprises from about 0.5 to about 3 wt.% high volatility compound.

41. The process for producing a polystyrene structure of claim 34 wherein said extrudate comprises from about 2 to about 10 weight percent blowing agent.

42. The process for producing a polystyrene structure of claim 41 wherein said extrudate comprises from about 3 to about 8 weight percent blowing agent.

10 43. The process for producing a polystyrene structure of claim 42 wherein said extrudate comprises from about 4 to about 6 weight percent blowing agent.

44. A blowing agent for foaming polystyrene consisting essentially of:
at least one low permeability compound, at least one of said low permeability
compound being selected from the group consisting cyclopentane, isopentane,
15 isobutane, 2-2 dimethyl butane, cyclohexane and mixtures thereof;

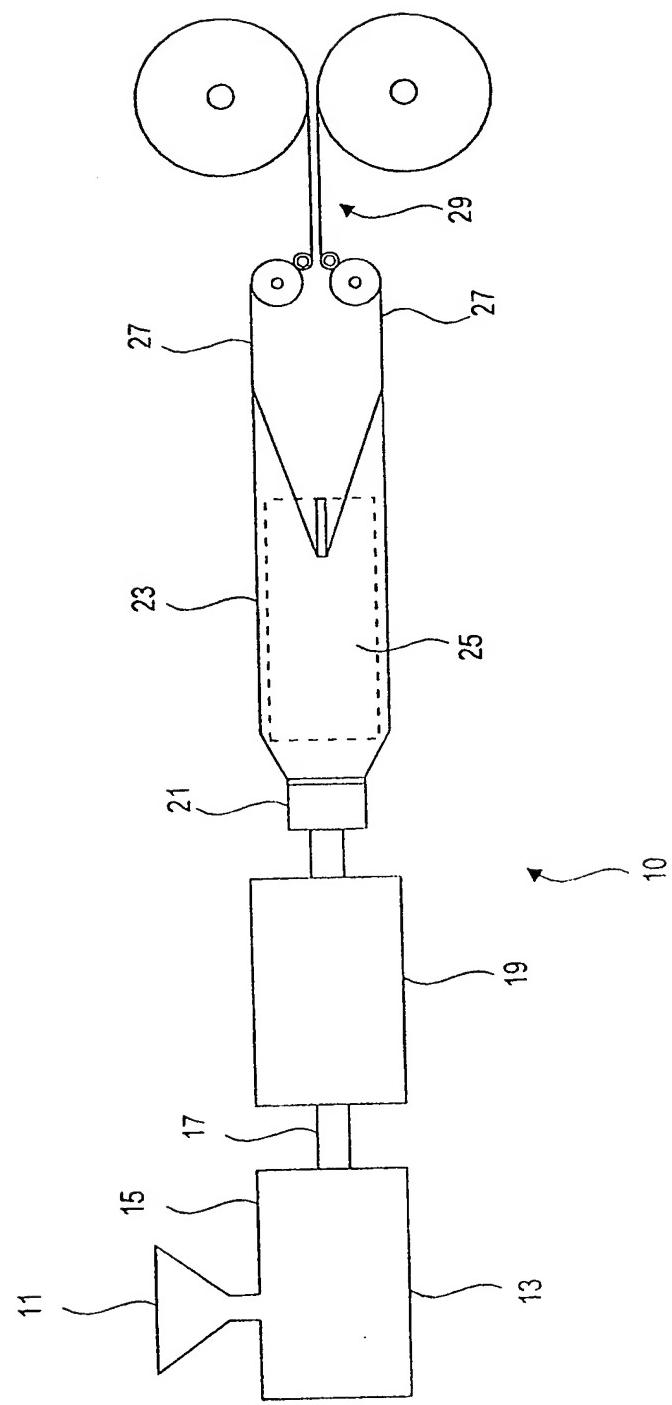
at least one low volatility compound, at least one of said low volatility
compound being selected from the group consisting of acetone, an acetone/water
mixture, methyl acetate and mixtures thereof; and

optionally, at least one high volatility compound, at least one of said high
20 volatility compound being selected from the group consisting of carbon dioxide,
nitrogen, hydrofluorocarbons, ethane and mixtures thereof;

wherein said blowing agent comprises less than about 42 weight percent low
permeability compound.

45. The blowing agent of claim 44 wherein at least one of said low permeability compound is selected from a group consisting of isopentane, isobutane and mixtures thereof, at least one of said low volatility compound is acetone and at least one of said high volatility compound is carbon dioxide.

5 46. The blowing agent of claim 44 wherein said blowing agent comprises less than about 34 wt.% low permeability compound.



INTERNATIONAL SEARCH REPORT

International application No.
PCT/US98/20507

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :C08J 9/04, 9/14; C09K 5/04
US CL :252/67, 69; 521/79, 97, 98, 130, 131, 133, 146

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 252/67, 69; 521/79, 97, 98, 130, 131, 133, 146

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant usages	Relevant to claim No.
X	US 5,453,454 A (ALICKE et al.) 26 September 1995, see the entire document.	1-45
A	US 5,576,094 A (CALLENS et al.) 19 November 1996, see the entire document	1-45
A	US 4,424,287 A (JOHNSON et al.) 03 January 1984, see the entire document.	1-45

Further documents are listed in the continuation of Box C.

See patent family annex.

* Special categories of cited documents	*T*	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
A document defining the general state of the art which is not considered to be of particular relevance		
E earlier document published on or after the international filing date	*X*	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
L document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*Y*	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
O document referring to an oral disclosure, use, exhibition or other means	*&*	document member of the same patent family
P document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search

08 DECEMBER 1998

Date of mailing of the international search report

31 DEC 1998

Name and mailing address of the ISA/US
Commissioner of Patents and Trademarks
Box PCT
Washington, D.C. 20231

Authorized officer

JOHN M. COONEY JR.

Faxsimile No. (703) 305-3230

Telephone No. (703) 308-0661

